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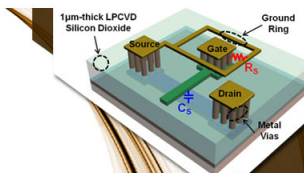
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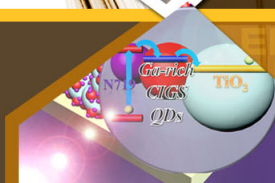
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# Integrated organic photovoltaic modules with a scalable voltage output

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Efficient organic photovoltaic modules and cells have been fabricated based on a blend of poly(3-hexylthiophene) (P3HT) and a soluble C<sub>70</sub> derivative, [6,6]-phenyl C<sub>71</sub> butyric acid methyl ester (PCBM-70). Power conversion efficiency of P3HT/PCBM-70 cells is estimated at 4.1% for the AM1.5 G condition from the spectra of external quantum efficiency. Integrated photovoltaic modules, in which *N*- individual P3HT/PCBM-70 cells (*N*=1–4) are connected in series, exhibit a short-circuit current *I*<sub>SC</sub> of 1.4±0.1 mA and an open-circuit voltage *V*<sub>OC</sub> of *N*×0.621 V with a maximum *V*<sub>OC</sub> of 2.48 V for *N*=4 under simulated solar illumination (AM1.5 G, 85 mW/cm<sup>2</sup>).

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Solar cells based on organic small molecules and conjugated polymers are being developed at a rapid pace in hope of thin, lightweight, and low-cost alternatives to conventional inorganic solar cells. While the current state-of-the-art organic photovoltaic (OPV) cells have demonstrated power conversion efficiencies around 4%-5%,<sup>1,2</sup> a study by Koster *et al.* suggests that even higher efficiency may be reached in an ideal donor/acceptor composition.<sup>3</sup> In order to demonstrate its full commercial potential, however, scalable OPV module technologies must be developed at the same time so that one can accommodate various needs of practical applications. Here, we report that efficient organic solar cells can be fabricated based on a mixture of poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl C<sub>71</sub> butyric acid methyl ester (PCBM-70), which was relatively rarely used when compared to the more common electron acceptor [6,6]-phenyl C<sub>61</sub> butyric acid methyl ester (PCBM-60).<sup>4</sup> Using the same mixture, we also demonstrate integrated OPV modules with a scalable voltage output and without short-circuit current reduction.

Discrete devices under study consist of an Al top electrode as a cathode and a bulk-heterojunction layer based on interpenetrating networks of P3HT and PCBM-60 or PCBM-70 that is spun on an indium tin oxide (ITO) anode coated with poly(3,4-ethylenedioxythiophene): poly(styrene sulfonate) (PEDOT:PSS). PCBM-60 and PCBM-70 were obtained from Solenne BV, and PEDOT:PSS (Baytron P VP Al 4083) was obtained from H.C. Starck. All of the chemicals were used as received without further purification. After spin-coating and a subsequent annealing (140 °C for 10 min) of PEDOT:PSS films,<sup>2</sup> samples were loaded into a nitrogen-filled glovebox, where a mixture of P3HT and PCBM in chlorobenzene (17 mg/ml) was spun at 700 rpm for 1 min. Then, those samples were brought into a vacuum deposition system (Spectros, Kurt J. Lesker) for deposition of Al electrodes. Typical device area for discrete photovoltaic (PV) cells was ~0.10±0.01 cm<sup>2</sup>. Samples were annealed on a hot plate at 150 °C for 30 min as was previously shown to

dramatically improve performance of P3HT:PCBM-60 cells.<sup>1,2</sup> Samples were not exposed to ambient air after being loaded into the glovebox, and all of the photovoltaic properties were measured in the glovebox unless noted otherwise.

Figure 1(a) shows electrical characteristics of devices with a geometry of ITO/PEDOT:PSS (26 nm)/P3HT:PCBM-60 or P3HT:PCBM-70 (1:1 by weight, 100 nm)/Al. Devices containing PCBM-70 (≡D70) exhibited a short-circuit current density *J*<sub>SC</sub> of 17.7±0.5 mA/cm<sup>2</sup>, when averaged for five devices, under the illumination of the broadband light from a filtered Xe lamp (CVI ASB-XE-175EX, 350–900 nm) having an irradiance *I*<sub>L</sub> of 100±2 mW/cm<sup>2</sup>. On the other hand, those with PCBM-60

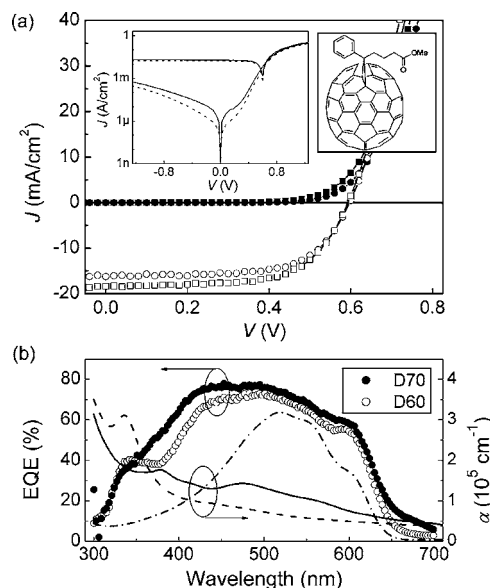


FIG. 1. (a) Electrical characteristics of the best devices having a geometry of ITO/PEDOT:PSS (26 nm)/P3HT:PCBM-70 (100 nm, 1:1 by weight)/Al (≡D70, squares) or ITO/PEDOT:PSS (26 nm)/P3HT:PCBM-60 (100 nm, 1:1 by weight)/Al (≡D60, circles) under illumination of the broadband light (100 mW/cm<sup>2</sup>, 350–900 nm, open shapes) and in the dark (closed shapes). Inset (left): Full-range curves in semilogarithmic scale under illumination and in the dark for D70 (solid) and D60 (dash). Inset (right): Molecular structure of PCBM-70. (b) External quantum efficiency (EQE) spectra of D70 (closed circles) and D60 (open circles). Also shown are absorption coefficients  $\alpha$  of PCBM-70 (solid line), PCBM-60 (dash), and P3HT (dash-dot) films obtained assuming Beer's law. The effect of reflection was neglected in estimation of  $\alpha$  from transmission measurements.

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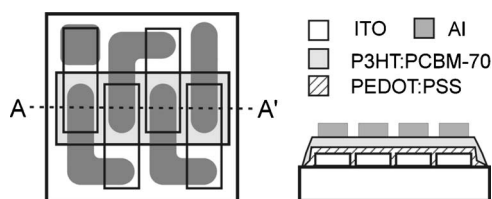


FIG. 2. Schematic device structure of an OPV module in which four individual cells are connected in series: top view (left) and cross-sectional view along the line AA' (right, not to scale). PEDOT:PSS is coated all over the substrate after ITO patterning, but not shown in the top view. The substrate size is approximately  $1 \times 1$  in.<sup>2</sup>.

(=D60) showed  $15.7 \pm 0.5$  mA/cm<sup>2</sup> on average, which is approximately 12% smaller than that of D70. Open-circuit voltage  $V_{OC}$  and fill factor FF were almost indistinguishable at  $593 \pm 9$  mV and  $0.63 \pm 0.01$  for D70 and  $594 \pm 3$  mV and  $0.65 \pm 0.01$  for D60. Overall power conversion efficiencies  $\eta$  ( $=FFJ_{SC}V_{OC}/I_L$ ) were thus  $(6.6 \pm 0.3)\%$  for D70 and  $(6.2 \pm 0.2)\%$  for D60 for the light source used for testing.

Figure 1(b) shows external quantum efficiency (EQE) as a function of wavelength  $\lambda$  for these devices (best one for each sample) measured at short-circuit condition. The higher EQE of D70 than that of D60 observed in most of the spectral region studied is consistent with higher  $J_{SC}$  values observed in D70. From the overlap integral of EQE spectra against the standard AM1.5 G condition ( $100$  mW/cm<sup>2</sup>),  $J_{SC}$  and  $\eta$  are estimated to be  $10.8$  mA/cm<sup>2</sup> and  $4.1\%$  for D70 and  $9.6$  mA/cm<sup>2</sup> and  $3.8\%$  for D60. Shown together in Fig. 1(b) are the absorption coefficients  $\alpha$  of individual films obtained using Beer's law with the effect of reflection assumed negligible. It shows that  $\alpha$  of PCBM-70 is higher than that of PCBM-60 in most of the spectral region,<sup>5</sup> which may be regarded as one of the main reasons for higher EQE values in P3HT/PCBM-70 cells. This appears to come from the similarity of PCBM-60 and PCBM-70 to their parent molecules C<sub>60</sub> and C<sub>70</sub> in the optical properties that are associated with a molecular symmetry, as discussed by Wien *et al.*<sup>4</sup> and Ren *et al.*<sup>6,7</sup> It is not yet clear whether the larger absorption coefficient of PCBM-70 is solely responsible for higher EQE values of PV cells than those of PCBM-60 based cells. Other factors such as internal morphology and balanced phase separation could also play key roles in carrier generation.

As a next step, we have fabricated OPV modules using P3HT/PCBM-70 blends<sup>8</sup> in a test geometry in which  $N$  individual cells are connected in series to produce a large voltage output as shown in Fig. 2. High, scalable voltage output is desired for many applications and is important especially when photovoltaic energy is used in conjunction with rechargeable batteries to initiate and maintain a charging condition. ITO bottom electrodes were patterned using a standard photolithography, and top electrodes were defined using a shadow mask with up to four active areas connected together. The active area of an individual cell in the modules is  $0.26$  cm<sup>2</sup>. The same PEDOT:PSS used for discrete cells was coated all over the substrate without patterning. PEDOT:PSS layers are expected to play some role in alleviating leakages and local shorts that can occur at sharp ITO boundaries by providing smoother interfaces. In-plane sheet resistance of the PEDOT:PSS layers used in this study was estimated to be on the order of  $10^9$   $\Omega$ /sq,<sup>9</sup> large enough to avoid cell-to-cell "cross-talk." The top and bottom parts of the P3HT/PCBM-70 composite films were wiped off after spin coating

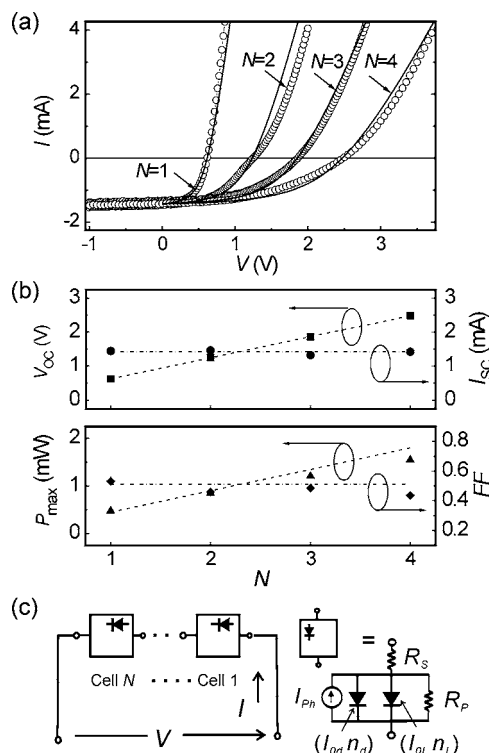


FIG. 3. (a) Electrical characteristics of OPV modules based on a blend of P3HT:PCBM-70 (3:2 by weight) consisting of a series connection of  $N$  individual subcells ( $N=1, 2, 3$ , and  $4$ ) measured in air under illumination from a 300 W solar simulator (AM1.5 G,  $85$  mW/cm<sup>2</sup>). (b) Scaling of PV parameters vs  $N$ :  $J_{SC}$  (circles),  $V_{OC}$  (squares),  $FF$  (diamonds), and  $P_{max}$  (triangles). The line associated with  $V_{OC}$  is the result of least-squared-error fit:  $V_{OC}(N)=N \times 0.621$  V. The solid lines in (a) and points at  $N=1, 2, 3$ , and  $4$  in all other lines of (b) indicate values predicted by PSPICE® simulation using an equivalent circuit model (ECM) shown in (c). In (b), line graphs were used for modeling results to better show trends in each case, although values are meaningful only when  $N$  is an integral number. In the subcircuit of the given ECM in (c), the diode represented by  $(I_{0L}, n_L)$  accounts for recombination of photogenerated carriers, which eventually modifies its  $I$ - $V$  shape from that of the "dark" diode represented by  $(I_{0d}, n_d)$ . See Ref. 12 for more details. Modeling results shown are for  $I_{ph}$  of  $1.46$  mA,  $(I_{0d}, n_d)$  of  $(3.1$  nA,  $1.98)$ ,  $(I_{0L}, n_L)$  of  $(12$   $\mu$ A,  $5.52)$ ,  $R_p$  of  $28400$   $\Omega$ , and  $R_s$  of  $50$   $\Omega$ . All parameters were first obtained from the best fit to a module with  $N=1$ , except for  $R_s$ , which was chosen so that fits to experimental  $FF$  and  $I$ - $V$  vs  $N$  have the least sum of squared errors.

to allow the metal electrode of one cell to be connected to the ITO electrode of an adjacent cell. Devices were encapsulated by putting UV-curable epoxy directly on the area with organic layers and by subsequently curing it under UV light for  $\sim 1$  h.<sup>10</sup> Testing was carried out in air under an AM1.5 G solar simulator (Oriel 91160,  $85$  mW/cm<sup>2</sup>) having a uniform light output across a  $2 \times 2$  in.<sup>2</sup> area.

Figure 3(a) shows electrical characteristics of modules with  $N=1-4$  that were fabricated from a P3HT:PCBM-70 blend ( $17$  mg/ml in chlorobenzene) with a 3:2 weight ratio. Note in Fig. 3(b) that  $V_{OC}$  scales linearly with  $V_{OC}(N)=N \times 0.621$  V and that short-circuit current  $J_{SC}$  remains almost constant at  $1.4 \pm 0.1$  mA, as can be expected from the general scaling rule for  $V$  and  $I$  in series connection. It should be noted that  $J_{SC}$  is limited by the cell producing the smallest photocurrent when there is a cell-to-cell imbalance within a series-connected module. Therefore, the fact that  $J_{SC}$  stays almost unchanged for different modules demonstrates a good cell-to-cell uniformity over the area of substrates used ( $\approx 1$  in.<sup>2</sup>) as well as a good sample-to-sample reproducibility.



Scaling of FF (and thus maximum available power  $P_{\max}$ ) vs  $N$  is not as trivial as  $V_{\text{OC}}$  and  $J_{\text{SC}}$ , in particular, in the presence of a series resistance  $R_S$  that increases with  $N$ . Increase of  $R_S$  with  $N$  can be easily noticed in Fig. 3(a) from the forward-characteristic slopes that decrease with  $N$ . Approximation of an  $N$ -cell module to an equivalent unit-cell device can give a simple insight into the scaling of FF.<sup>11</sup> That is, when the shunt resistance is large enough for its effect to be negligible, the net fill factor of an  $N$ -cell module  $\text{FF}(N)$  can be approximated by

$$\text{FF}(N) \approx \text{FF}_0(N)[1 - r_S(N)], \quad (1)$$

in which  $\text{FF}_0(N)$  is the net ideal fill factor of an  $N$ -cell module that has no series resistance and  $r_S(N)$  is the net normalized series resistance defined by  $R_S(N)I_{\text{SC}}(N)/V_{\text{OC}}(N)$ . From the general scaling rule for  $V$  and  $I$ ,  $\text{FF}_0(N) = \text{FF}_0(N=1)$ ,<sup>11</sup>  $I_{\text{SC}}(N) = I_{\text{SC}}(N=1)$ , and  $V_{\text{OC}}(N) = NV_{\text{OC}}(N=1)$ . Note that  $R_S(N) = NR_S(N=1)$  if  $R_S(N)$  results from replication of  $R_S$  of individual cells and if they are all same. In such case, one can easily see that  $\text{FF}(N)$  would also remain unchanged at  $\text{FF}(N=1) = \text{FF}_0[1 - r_S(N=1)]$  and that  $P_{\max}$  ( $= \text{FF}I_{\text{SC}}V_{\text{OC}}$ ) would increase linearly with  $N$ . Such scaling of  $P_{\max}$  indicates that effective power conversion efficiency  $\eta_{\text{eff}}$  defined for the active device area  $A(N)$  of  $N \times 0.26 \text{ cm}^2$  would stay constant regardless of  $N$ .

More advanced PSPICE® simulation using an equivalent circuit model<sup>12</sup> (ECM) shown in Fig. 3(c) not only demonstrates that the experimental  $I$ - $V$  characteristics can be well fitted to the given ECM [see lines in Fig. 3(a)] but also confirms the trend of FF and  $P_{\max}$  that is predicted by the above simplified argument [see lines in Fig. 3(b)]. Actual experimental  $P_{\max}$ , however, increases in a slightly sublinear fashion as can be seen in Fig. 3(b) and actual FF (and thus  $\eta_{\text{eff}}$ ) decreases with  $N$  from 0.53 (2.2%) for  $N=1$  to 0.44 (1.7%) for  $N=4$ . This may be attributed to cell-to-cell variations in  $R_S$  and other equivalent circuit parameters that can cause nonideal  $I$ - $V$  characteristics. Future work will require design improvements that minimize the series resistance of individual subcells and interconnections. In addition, dense packing of individual subcells should be realized for maximal utilization of a substrate area. To achieve that, however, one should develop a method that allows precise patterning or placement of organic and metal layers, without compromising device performance or adding significant cost. Such a study is currently under way, and also Dennler *et al.* have reported their efforts to introduce a laser-etching technique

used in fabrication of monolithically integrated  $a$ -Si PV modules to OPV.<sup>13</sup>

In summary, we demonstrated that efficient organic solar cells can be achieved using a bulk heterojunction of P3HT and PCBM-70. We also demonstrated that integrated OPV modules with a scalable voltage output can be fabricated based on this P3HT/PCBM-70 blend. Although the current module design still has room for improvement, we believe that these results are an important step forward toward a practical use of OPV technologies.

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<sup>5</sup>In the UV range, one can observe an absorption peak in PCBM-60 films at 3.7 eV, but this peak is less critical for solar power generation because the sunlight has only small power contents in this range.

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<sup>8</sup>P3HT used for discrete PV cells was from R. D. McCullough at Carnegie Mellon University. Due to its limited quantity, P3HT used for fabrication of modules was purchased from Rieke Metals.

<sup>9</sup>The grade used here has a conductivity  $\sigma$  of  $10^{-3} \text{ S/cm}$ . Some high-conductivity grade PEDOT:PSS with  $\sigma > 100 \text{ S/cm}$  could be problematic as the sheet resistance can become on the order of  $\text{k}\Omega/\text{sq}$  even for 10–100 nm thick films.

<sup>10</sup>It was shown in an independent experiment that this encapsulation method could provide adequate protection for a short period of time for testing, although it can still suffer from degradation to a certain degree [W. J. Potscavage, S. Yoo, X. H. Zhang, X. D. Wang, B. Domercq, Z. L. Wang, and B. Kippelen (in preparation)].

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